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TITLE: RECENT DEVELOPMENTS AT LOS ALAMOS FOR THE
MEASUREMENT OF ALPHA CONTAMINATED WASTE

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RECENT DEVELOPMENTS AT LOS ALAMOS FOR THE MEASUREMENT OF ALPHA CONTAMINATED WASTE

ABSTRACT

A Comprehensive program is currently in progress at the Los Alamos Scientific Laboratory dedicated to the development of sensitive, practical nondestructive assay techniques for the quantification of low level transuranics in bulk solid wastes. This program encompasses a broad range of nuclear and non-nuclear techniques including sophisticated passive gamma-ray and passive neutron detection systems, isotopic neutron source based active interrogation systems, pulsed portable neutron generator active interrogation systems, electron accelerator based techniques and laser spectroscopy techniques. The mix of techniques ranges in development maturity from the well established (MEGAS, Shuffler, Passive 4π neutron counters) through the proof-of-principle stage (pulsed neutron generator techniques) to the under investigation stage (electron linac and laser spectroscopy techniques). The program is intended to provide technical solutions for waste management problems of today as well as of tomorrow. We are concentrating on specific measurement problems associated with different waste matrices and are developing matrix compensation methods to improve the accuracy of waste screening and assay measurements.

Specific detection systems have been designed to operate in the high level beta-gamma backgrounds associated with some commercial reactor wastes. The techniques being developed can be used with either low level or high level beta-gamma wastes in either low density or high density matrices. Engineering criteria provided by waste generating facilities are an important feature in our design efforts.

1. INTRODUCTION

A vital part of any comprehensive radioactive waste program is the ability to determine the types and quantities of radioactivity present. Without this ability, it is impossible to direct a cost-effective and timely waste management program in any phase of the nuclear industry. Assay or verification capability also provides quality assurance for operating health and

safety aspects of the facility involved. The Los Alamos Scientific Laboratory (LASL) is developing nondestructive assay (NDA) techniques for the analysis of transuranic (TRU) isotopes found in bulk solid wastes generated in U.S. governmental and commercial nuclear industries. Some of the equipment and techniques under development have application to non-TRU wastes as well. All techniques are being developed so as to be useable with high beta-gamma TRU wastes as well as with "quiet" wastes.

The program at LASL can be divided naturally into three broad categories:

Category I.

The engineering development of established TRU measuring techniques, specialized to meet specific waste management needs according to sensitivity and waste package characteristics.

Category II.

The development of new and potentially more sensitive and/or useful TRU measuring techniques that have reached a proof of principal level.

Category III.

The investigation of novel measurement techniques that have the potential of characterizing TRU waste accurately and sensitively according to each isotope present, for which the proof of principal measurements do not yet exist.

The LASL program thus features technique and equipment development spanning a large range of development maturity. In Category I, we place such established techniques as the Multi-Energy Gamma Assay System^[1] (MEGAS), which is based on collimated passive gamma scanning of waste packages. This system is intended for assay/screening at the 10 nCi/g level of low density waste packages. A number of these systems are in use for particular waste assay applications at LASL and other laboratories. Significant improvements^[2-4] in this system have been made recently.

Another technique falling in Category I is the "²⁵²Cf Shuffler."^[5] This technique has been used for various purposes in the nuclear industry and a system is now under engineering development at LASL,^[6] designed specifically for assay/screening of high density 200-liter waste packages at the 10 nCi/g level. The technique is based on cyclical neutron irradiation of waste barrels with a ²⁵²Cf source, which can be stored in a highly shielded location during the subsequent induced-fission-delayed-neutron counting portion of the cycle. This measurement directly determines fissile content. The delayed neutron detection system can also be used for passive neutron coincidence counting of 200-liter waste barrels, which

quantifies the spontaneous fission emitters in the waste. The shuffler system is capable of TRU analysis even in the presence of very high beta-gamma levels,^[6] such as from commercial reactor wastes.

We are currently developing a third Category I system. This system will perform a highly sensitive 4π solid angle passive neutron count of very large TRU waste packages having external dimensions of 1.2 m x 1.2 m x 2.1 m. This system is based on a flexible, modular detector design allowing for the easy construction of even larger (or smaller) 4π solid angle counting systems using the same detector modules. This system is intended for the assay/screening at the 10 nCi/g level of large bulk waste storage containers ("crates") commonly used in the U.S. and for the assay/screening of aggregates of smaller containers such as the "6 pack" of 200-liter barrels also in common use in the U.S.

A very promising Category II system under development is called the Differential Dieaway Technique (DDT) and is based on pulsed neutron interrogation. A prototype 200-liter waste barrel assay system is currently in use with a portable 100-pulse per second (pps) 14-MeV neutron generator as the interrogating source. Very high sensitivity for ^{235}U , ^{239}Pu , and ^{241}Pu in bulk wastes is obtained by counting the induced prompt fission neutrons after each pulse. This system is intended for use on low level TRU (1-10 nCi/g or less) bulk waste assay/screening problems.

Our Category III techniques are currently all associated with our linear electron accelerator based program. This work is covered in some detail in an accompanying paper.^[7] In this program a variety of techniques is being investigated in the hope of achieving specific multi-isotope assay capability. Prompt photofission coincidence neutron counting, photo- and neutron-induced fission delayed neutron counting, and energy differential photofission yield techniques are among those being pursued. In addition, a novel technique based on isotope specific fission product noble gas assay using the laser based resonance ionization spectroscopy (RIS) technique is under investigation.^[8,9] In this approach specific TRU isotopes would be identified and quantified based on differential yields of specific noble gas (Xe and Kr isotopes) fission products.

2. WASTE MATRIX STUDIES

All the specific techniques discussed in Section I can yield reasonably accurate TRU assay values. However, all the techniques can also be subject to serious measurement errors in unfavorable waste matrices. A considerable portion of our development efforts go into waste matrix compensation studies;

that is, to the development of methods for compensating in the data analysis for errors produced by the presence of the non-TRU materials in bulk wastes.

In our passive x-ray and gamma counting systems, the principal sources of matrix generated assay errors are gross gamma-ray scattering and/or attenuation and, for some high TRU wastes, self-attenuation. For most practical screening/ low precision assay applications, matrix effects are adequately taken into account with calibrations based on standards placed in typical matrix geometries. A more sophisticated compensation is based on an adjunct external gamma source transmission measurement of the waste container from which an average gamma-ray attenuation factor may be derived. In any event, the accuracy of passive gamma-ray counting is generally quite good for low density wastes (density $< 0.2 \text{ g/cm}^3$), but is poor for higher density wastes.

In high density TRU waste assay systems involving neutron counting, the principal matrix associated errors are caused by hydrogen scattering/attenuation and thermal neutron absorption. A well designed counter can achieve a considerable degree of intrinsic matrix compensation. For example, ^3He proportional counters with about 2.5 cm of polyethylene moderator in front and > 2.5 cm of polyethylene in back exhibit essential flatness of response for matrix hydrogen densities ranging from zero to the equivalent of about 3-cm thickness of water moderator.

Another matrix hydrogen density compensation technique uses the so-called "ring-ratio" or "slab-ratio" method. Here the detector is constructed so as to provide neutron spectral information. This spectral information can then be interpreted in terms of equivalent hydrogen density in the waste matrix and appropriate corrections can be made in the assay analysis.

In the pulsed active neutron techniques we have been able to achieve matrix compensation using thermal neutron interrogation cavity flux monitors. Interrogating flux reductions or increases produced by matrix constituents are measured by the flux monitor. In addition, we have been able to measure interrogation cavity thermal neutron lifetimes with this system and have found a strong correlation between average neutron lifetime and the amount of neutron absorbing materials in the matrix. We are currently evaluating this technique for routine use. Another approach under investigation uses the measurement of matrix capture gamma-ray spectra to determine matrix composition. This measurement is done simultaneously with the induced fission neutron measurement.

3. CATEGORY I TECHNIQUES

3.1. Improvements in the MEGAS system

The basic operation of the original MEGAS system is described in Ref. [1]. Recently, the original MEGAS has been considerably upgraded (MEGAS II). [2-4] The original NaI detector has been replaced with a 127-mm diameter x 1.6-mm thick NaI crystal, which optimizes TRU detection capability using L x-rays and gamma rays of less than 100 keV. The thinner crystal also results in a decreased Compton scattered background. With this detector, the detection limit at the 3σ level above background for ^{241}Am is less than 5 pCi/g for a 500-s count for approximately 6 kg of low density wastes in a 57-liter carton.

The presence of beta- and gamma-ray emitting fission products severely decreases the achievable TRU detection limit for even thin NaI detectors. However, the recent addition [3,4] of a high resolution hyperpure planar germanium detector, 1000-mm² active area, 12 mm thick allows us to assay TRU isotopes even in the presence of considerable extraneous gamma and x-ray backgrounds.

A tabulation of measured detection limits for the hyperpure germanium detector over a wide range of photon energies is presented in Ref. [3]. Using these data, we estimate that TRU assay at the 10 nCi/g level can still be made even in the presence of 400 μCi of ^{137}Cs (65 nCi $^{137}\text{Cs/g}$).

We have also added four banks of polyethylene moderated ^3He neutron detectors to MEGAS II. The measured detectability limit for this neutron detection system is 10 mg of typical reactor grade plutonium oxide. This detection limit applies for a net signal 3σ above background for a 1000-s run time using the total neutron count. [2,3] Because ^3He detectors are relatively insensitive to photons, this part of the system can detect TRU-related neutrons in a container even in the presence of high fission product backgrounds (1-10 R/h).

MEGAS II operates in a segmented scanning mode, which allows one to locate hot spots within waste packages. This information is useful in determining whether individual waste containers should be repackaged or material recovered.

3.2. The ^{252}Cf Shuffler system

The ^{252}Cf Shuffler system combines active and passive neutron assay techniques to measure plutonium contaminated waste and is described in detail in Refs. [5] and [6]. The key features of the technique include an assay chamber large enough to accommodate a standard 200-liter barrel surrounded by ^3He filled proportional gas detectors for efficient neutron counting and

a 2-mg ^{252}Cf neutron source that can be used to interrogate the waste or can be stored in a shielded position, which is decoupled from the assay chamber.

A typical 1000-s assay begins with a passive neutron measurement with the source in the storage position. The passive count measures the total neutron emission rate and the coincidence rate arising from spontaneous fission of the even plutonium isotopes. The detectability limit for total neutron passive counting for 200-liter barrels of high density TRU ranges from 0.3-30 nCi/g depending on the matrix chemical composition.^[10] The coincidence technique, which provides an accurate assay independent of matrix chemical composition, displays a detection limit of about 10 nCi/g if no lead shielding is required. The same detection limit is obtained even with lead in place if adequate cosmic-ray shielding is provided.^[10]

The active portion of the assay is cyclical with repeated neutron irradiation and delayed neutron counting. One active cycle consists of a 16-s neutron interrogation and an 8-s delayed neutron count with the source withdrawn. The source transfer of about 1.8 m requires 0.5 s in either direction for a total cycle time of 25 s. A maximum of 30 cycles is used in the active portion of the assay.

In order to permit the active technique to be used for high-level, beta-gamma contaminated waste, a 15-cm-thick lead shield is placed between the barrel and neutron detectors. The shield permits operation in gamma radiation fields as high as 1000 R/h. With a nominal 1-m thick concrete cosmic-ray shield, the calculated 3 σ detectability limit with lead in place and assuming the use of a 2 mg ^{252}Cf interrogating source is 4.2 mg plutonium (4.6 nCi/g for a barrel with a 100-kg total mass containing 20% plutonium grade TRU). Combining the coincidence and integral passive counting data with the active assay results yields the fertile and fissile plutonium components as well as the probable average chemical composition.

3.3. A high efficiency passive neutron counting system for very large transuranic waste packages

In the United States a very common TRU waste storage and transport package is a wooden box (called a "crate") having external dimensions of 1.2 m x 1.2 m x 2.1 m. The large physical size of such packages has in the past presented great difficulties for TRU assay and/or screening measurement systems. In addition, it is common practice at many facilities producing TRU waste to band together six of the common 200-liter waste barrels into a large "six pack" package for convenience in transportation and/or storage. The external dimensions of these waste packages are typically 1.2 m x 1.0 m x 1.9 m, which are again very difficult to assay or screen for TRU activity with currently existing equipment.

We have embarked on a detector development program aimed specifically at this very large waste package assay/screening problem. The approach we are pursuing is a 4π solid angle passive neutron counting system sufficient in size to accommodate either of the common large waste packages mentioned above. Because facilities generate even larger packages, we have pursued a modular approach in the detector design so that a larger (or smaller) 4π detection system may easily be built using the same modules. In fact, in the recent past we have built similar 4π neutron counting systems of dimensions up to 2.4 m x 3.0 m x 6.1 m, intended for use as vehicle portal monitors. [11]

The principal isotope producing neutrons in common TRU waste is ^{240}Pu , although, depending on the chemical form, all alpha-emitting isotopes in the waste contribute to the neutron output of the package. In our system two types of passive neutron measurements are carried out simultaneously: total neutron output and coincidence neutron rate. The latter is a direct measure of the spontaneous fission rate of all fissioning materials in the waste package, whereas the former depends, in addition, on all alpha-emitting material in the package and on the chemical form of the waste.

In typical light water reactor fuel waste (assuming Cm isotope removal), the total neutron source for TRU waste in the oxide form is about 300 n/s per gram of included TRU. This will increase by about 10% in five years with the grow-in of ^{241}Am . The inclusion of certain low atomic number materials, such as Be, B, Li, F, etc., will also lead to an increased source term. The spontaneous fission neutron source for this waste is on the order of 200 n/s per gram of included TRU, which corresponds to about 100 spontaneous fissions per second per gram of included TRU.

The basic module in our system is a rectangular cross section (2.1 m x 1.2 m) polyethylene (CH_2) box in which are placed several 1.82 m x 0.05 m x 2 atm fill pressure ^3He proportional counters. We have found that this "neutron chamber" approach makes very effective use of ^3He proportional counters as compared to the usual practice of imbedding proportional counters in a solid CH_2 matrix and also results in reasonably portable modules. Our measured intrinsic neutron detection efficiency for this module for a bare ^{252}Cf spontaneous fission source is 16% when six 1.82 m x 0.05 m x 2 atm ^3He proportional counters are placed with equidistance spacings within the CH_2 neutron chamber. The same module displays a 15% intrinsic detection efficiency for a ^{252}Cf source moderated by 3 cm of CH_2 .

Optimization studies have revealed that improved performance per ^3He counter is obtained with a two-chambered module. For instance, a module constructed of 1.2-cm thick

CH₂ with a 2.5-cm thick CH₂ dividing wall displays an intrinsic ²⁵²Cf spectrum neutron efficiency of 20% with the six ³He counters divided between the two chambers. An added feature of this arrangement is that the count rate ratio between the two chambers is spectrally sensitive and may be used to correct for waste matrix generated efficiency modifications.

As pointed out in Ref. [11], a large increase in detection sensitivity in 4 π neutron counters is obtained by shielding out cosmic ray generated backgrounds. We propose taking advantage of this fact by providing an economic compacted dirt shield for our system. The housing for such an economic system can be made from prefabricated 3-m diameter (or larger) concrete drainage culvert piping, which are typically rated for > 5-m compacted dirt overburden. Such a 5-m dirt shield, easily implemented at waste burial or storage grounds, would result in a factor of 10 improvement in detection sensitivity as compared to the unshielded case. The cosmic-ray shielding factor for this system is about 100.

The current version of our very large package counting system has a 4 π intrinsic detection efficiency of 15-20%, depending on the modules used. We plan to use eight of the modules discussed earlier, routing separate inputs to a micro-processor. Our calculated system minimum detection sensitivity for a typical TRU isotopic mix resulting from light water reactor generated waste is about 10 mg of included TRU. This assumes extensive cosmic ray shielding, a 1000-s count time, and is based on obtaining a net coincidence count 3 σ above background. The 10-mg sensitivity applies to TRU located anywhere within a 1.2 m x 1.2 m x 2.1 m waste package.

4. CATEGORY II TECHNIQUE--A 1-nCi/g SENSITIVITY TRANSURANIC WASTE ASSAY SYSTEM USING PULSED NEUTRON INTERROGATION--THE DIFFERENTIAL DIEAWAY TECHNIQUE

Over the past several months we have developed a sensitive low-level transuranic waste assay system designed primarily for the rapid screening or assay of high density wastes contained in 200-liter steel barrels.[12,13] Our system is based on the principle of pulsed thermalized neutron interrogation with detection of induced fission prompt neutrons. This approach leads to greatly enhanced sensitivity as compared to techniques using the detection of delayed neutrons. In the case of ²³⁹Pu neutron fission, for example, the ratio of prompt fission neutrons to delayed fission neutrons is about 500.

Our prototype assay system has a square cross section assay chamber, which is 75 cm on a side, 95-cm high, lined with a 10 cm thick layer of graphite, and easily accommodates a standard 200-liter barrel. (A schematic drawing of this system is shown in Fig. 1.) Behind the graphite is a 15-cm thick layer of poly-

ethylene placed for internal neutron reflection and external personnel shielding. Our neutron source is a portable, light-weight (10-cm o.d., x 30-cm long x 10 kg), D+T pulsed neutron generator (100 pps, 10 μ s pulse width, 1×10^6 n/pulse). The neutron generator is positioned within the interrogation cavity. A pulse of 14-MeV neutrons is rapidly moderated and thermalized within the graphite and polyethylene. Following thermalization, neutrons diffuse from the graphite into the chamber, interrogating the waste barrel over an effectively stretched out time period of several milliseconds. (The observed interrogation chamber neutron time history is shown as Curve b in Fig. 2.) We observe a thermal neutron dieaway time of 1.3 ms for the empty cavity and 0.76 ms when a 200-liter barrel filled with 200 kg of sand and vermiculite is placed within the cavity.

To detect the induced prompt fission neutrons, we place moderated ^3He proportional counters between the graphite and polyethylene layers. These operate functionally as fast neutron detectors, having a minimal amount of moderator around them (about 1-cm average thickness) and are heavily shielded from any incident thermal neutrons with layers of boron and cadmium. This combination results in a rapid recovery from the initial pulse overload--no neutrons from the initial 14-MeV pulse are detected after a 0.5-ms time period. (The neutron time history for this detector is shown in Fig. 2, Curve a.) However, if a thermal neutron induced fission occurs within the interrogation cavity after 0.5 ms, the ensuing prompt fission neutrons propagate to the fast neutron detector and may be recorded. Thus, effectively, our "external" fast neutron detector is decoupled from the interrogating thermal neutron flux (after 0.5 ms) but is sensitive to the prompt fast neutrons accompanying an induced fission reaction.

We have an additional bare low ^3He fill pressure internal flux monitor detector located within the assay chamber. This detector allows us to compensate in the assay analysis for any pulse-to-pulse variation in the neutron generator output and for interrogating flux perturbations produced by the barrel or its contents.

We have performed extensive tests using a 200-liter barrel filled with 200 kg of sand and vermiculite to simulate a typical high density waste matrix. Aluminum tubes were placed in the barrel at its center, mid-radius, and near its outer periphery to facilitate placement of test samples throughout the volume of the barrel. Our external fast neutron detection system consists of three, 90 cm x 5 cm x 2 atm, moderated ^3He proportional counters. With this simple system we routinely detect 3-mg ^{235}U samples at the 3σ above background level in 8000-pulse runs with the ^{235}U sample placed at the center of the barrel full of sand. (Yield data as a function of ^{235}U mass are shown in Fig. 3. These data were taken with low ^{235}U enrichment uranium samples.) This is 80 seconds of elapsed time with the

neutron generator operating at its nominal 100-pps rate. As can be seen in Fig. 3, we observe a linear assay response for our system. We also have observed with the 200-liter barrel of sand in the cavity a reasonably flat ($\pm 10\%$) interrogation flux profile within the barrel.

Because our system is 1.5 times more sensitive to ^{239}Pu than to ^{235}U , the 80-s interrogation, 3σ detection limit for ^{239}Pu is 2 mg for the prototype system in its present configuration. Two milligrams of plutonium in a 200-liter barrel filled with 200 kg of sand is about 0.8 nCi/g matrix. Thus, we have demonstrated with a very simple and relatively inexpensive system (neutron generator with all associated hardware costs less than \$25k) a sub-1 nCi/g detection sensitivity capability for 200-liter barrels containing high matrix density TRU waste.

Recently we demonstrated that the DDT system could be used with $\text{Be}(\gamma, n)$ neutrons produced by pulsed low energy electron bremsstrahlung incident on a portion of the graphite cavity wall that had been replaced with Be. The EG&G/Santa Barbara electron linac was used for this purpose and both 6- and 8-MeV bremsstrahlung were used successfully. In fact, the interrogating neutron flux produced by a single 15 mA, 4 μs duration electron burst (typically used for these measurements) is some 3-5 times more intense than that produced by a single burst from the pulsed D+T generator described earlier.

In all respects the fissile assay done with the DDT system using a pulsed $\text{Be}(\gamma, n)$ interrogating source was identical to that obtained with the D+T neutron generator. As part of the $\text{Be}(\gamma, n)$ interrogating source measurements, we did systematic fissile assay measurements with a set of PuO_2 samples ranging in Pu mass from 1 to 1000 mg. These were successively placed at the center of a 200-liter barrel inside the DDT interrogating cavity, with measurements made for four different sets of typical matrix materials filling the 200-liter barrel. The "typical" matrices included high density ($>0.8 \text{ g/cm}^3$) barrel fillings of a) sand and vermiculite, b) aluminum scrap, c) aluminum scrap and polyethylene, and d) aluminum scrap, polyethylene, and borated glass beads.

We found that to an accuracy of about $\pm 20\%$ we obtained a matrix independent assay for all four matrices, and, in addition, for samples placed within the empty cavity. For these measurements the internal ^3He flux monitor was used to normalize to a constant number of effective interrogating neutrons. This matrix independence is remarkable considering the perturbation of the interrogating flux by the matrix. We measured independently the number of $\text{Be}(\gamma, n)$ neutrons produced for each measurement and, based on these data, found that for matrices a), b), and c), the effective interrogating flux was reduced by a factor of two compared to the empty cavity case and for matrix

d) was reduced by a factor of four. However, by the simple use of the internal flux monitor all these measurements yielded the same value on a normalized basis.

For a 200-s linac interrogation run our 3σ detection limits ranged from 0.4 to 0.7 mg Pu, depending on the matrix in the barrel. This corresponds to a detection limit well under 1 nCi/g for all the matrices tested. Figure 4 shows a typical set of this data.

Thus, we have demonstrated that the DDT system can be used successfully with either a pulsed D+T neutron generator source or with a low energy electron bremsstrahlung Be(γ ,n) source. The former (small pulsed D+T source) is available from commercial suppliers at an estimated cost of \$25k and the latter (medical or industrial x-ray linac) is also available commercially at an estimated cost of \$300k.

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6. FIGURE CAPTIONS

- Fig. 1. Schematic drawing of the prototype Differential Dieaway Technique Interrogation Cavity and Detection System. The current version of this system has external neutron detectors located on all four vertical walls and four independent flux monitors positioned in the interrogation cavity.
- Fig. 2. Pulsed neutron time history data for (a) external detection system and (b) interrogation cavity. This shows results obtained with a 200-liter barrel of sand in the interrogation cavity and no fissile material present. Introduction of fissile material results in an additional component in curve (a) having $T_{1/2} \sim 0.76$ ms and attributable to prompt fission neutrons.
- Fig. 3. Experimental response obtained with the prototype system shown in Fig 1. for 8000 interrogating pulses (80 s) from a 14 MeV neutron generator having a nominal 1×10^6 neutrons per pulse output. Low enrichment ^{235}U samples were placed at the center of a 200-liter barrel filled with sand for these measurements. The observed 3σ detection limit is 3 mg of ^{235}U .
- Fig. 4. Experimental response obtained in a series of 200-s runs with the Differential Dieaway Technique system (Fig. 1.) using pulsed $\text{Be}(\gamma, n)$ neutrons as the interrogating source. Data shown are for 20,000 electron linac pulses with about 4×10^6 $\text{Be}(\gamma, n)$ neutrons produced per pulse. Run total interrogating flux is about ten times that for the data shown in Fig. 3. Samples of PuO_2 were placed at the center of a 200-liter barrel of sand. The observed 3σ detection limit is 0.4 mg Pu (~ 0.2 nCi/g).

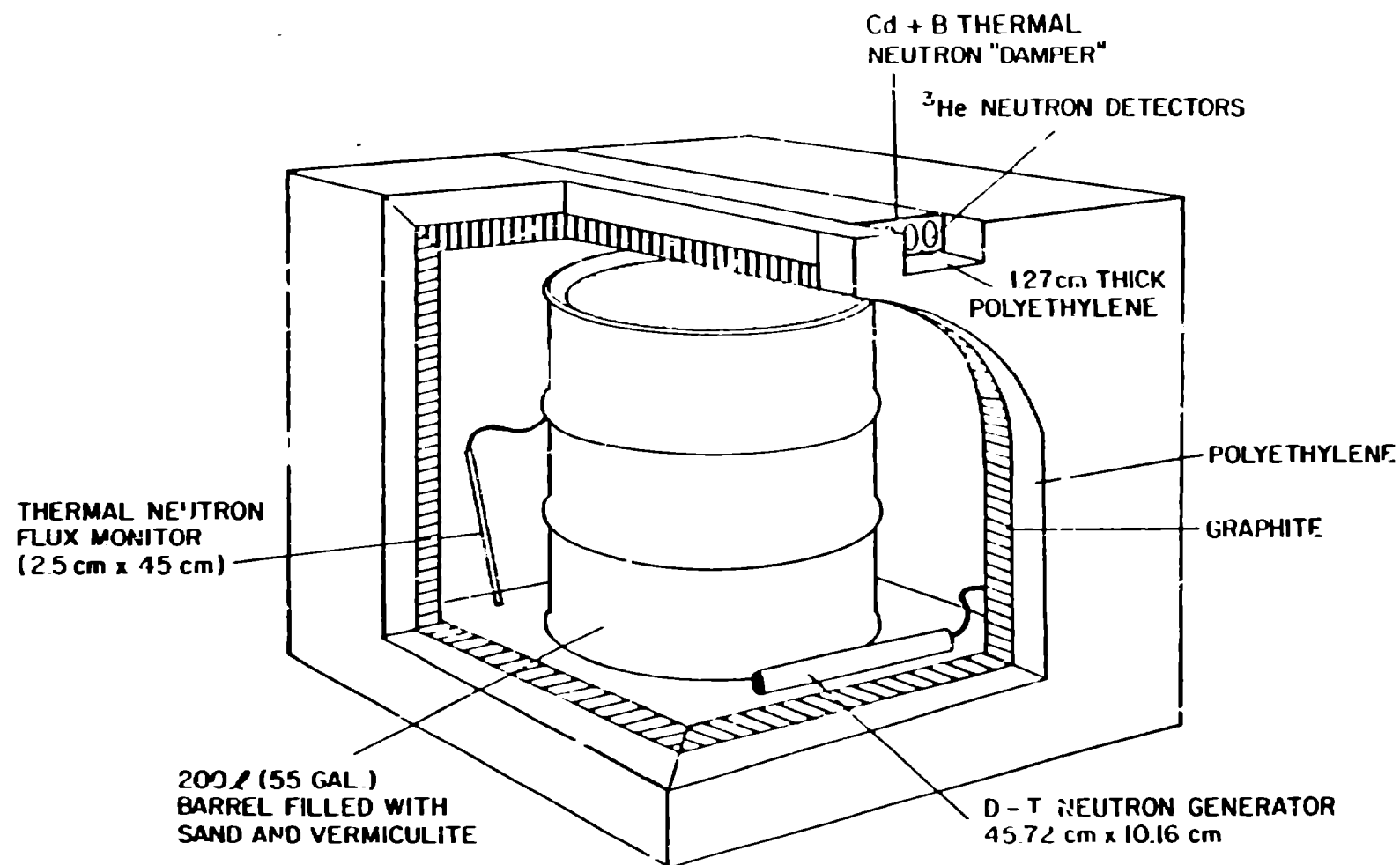


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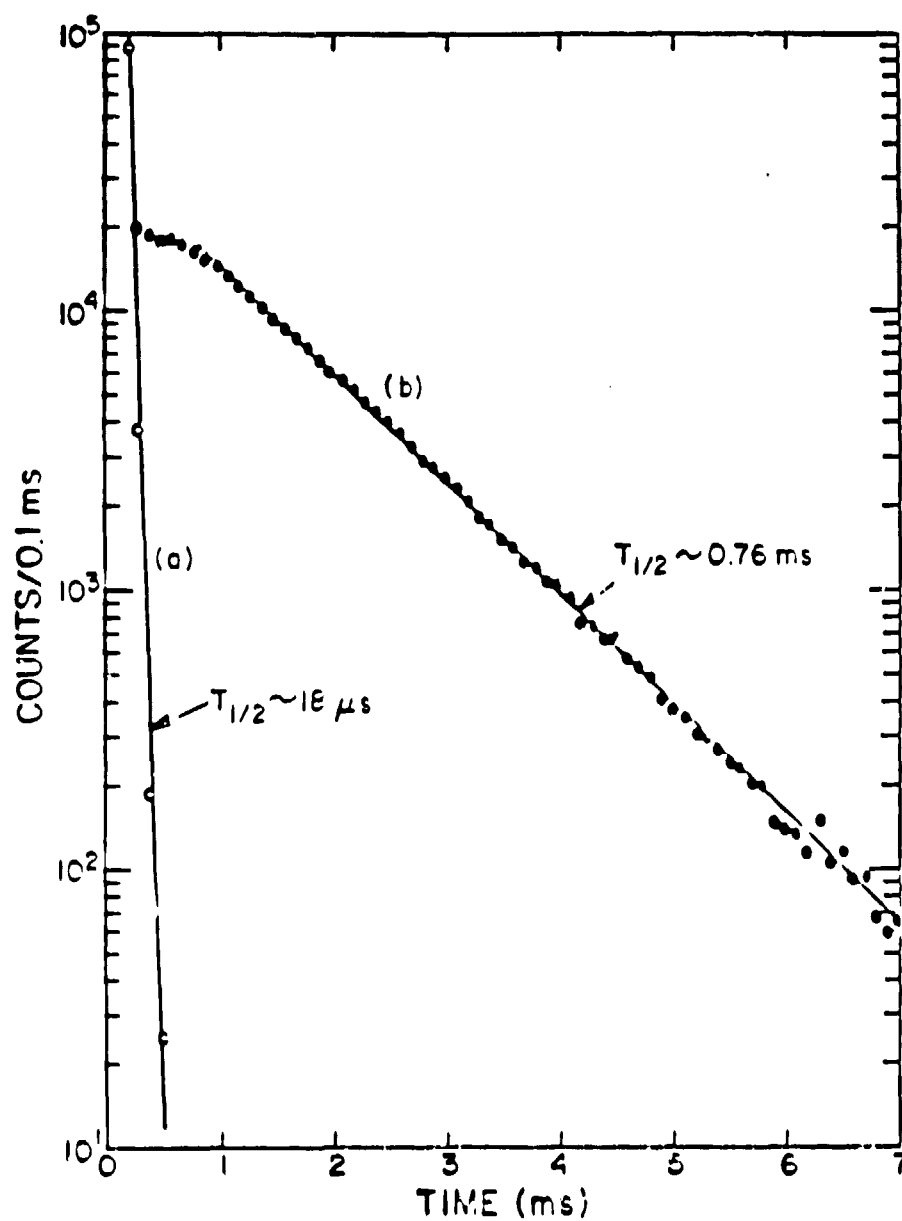


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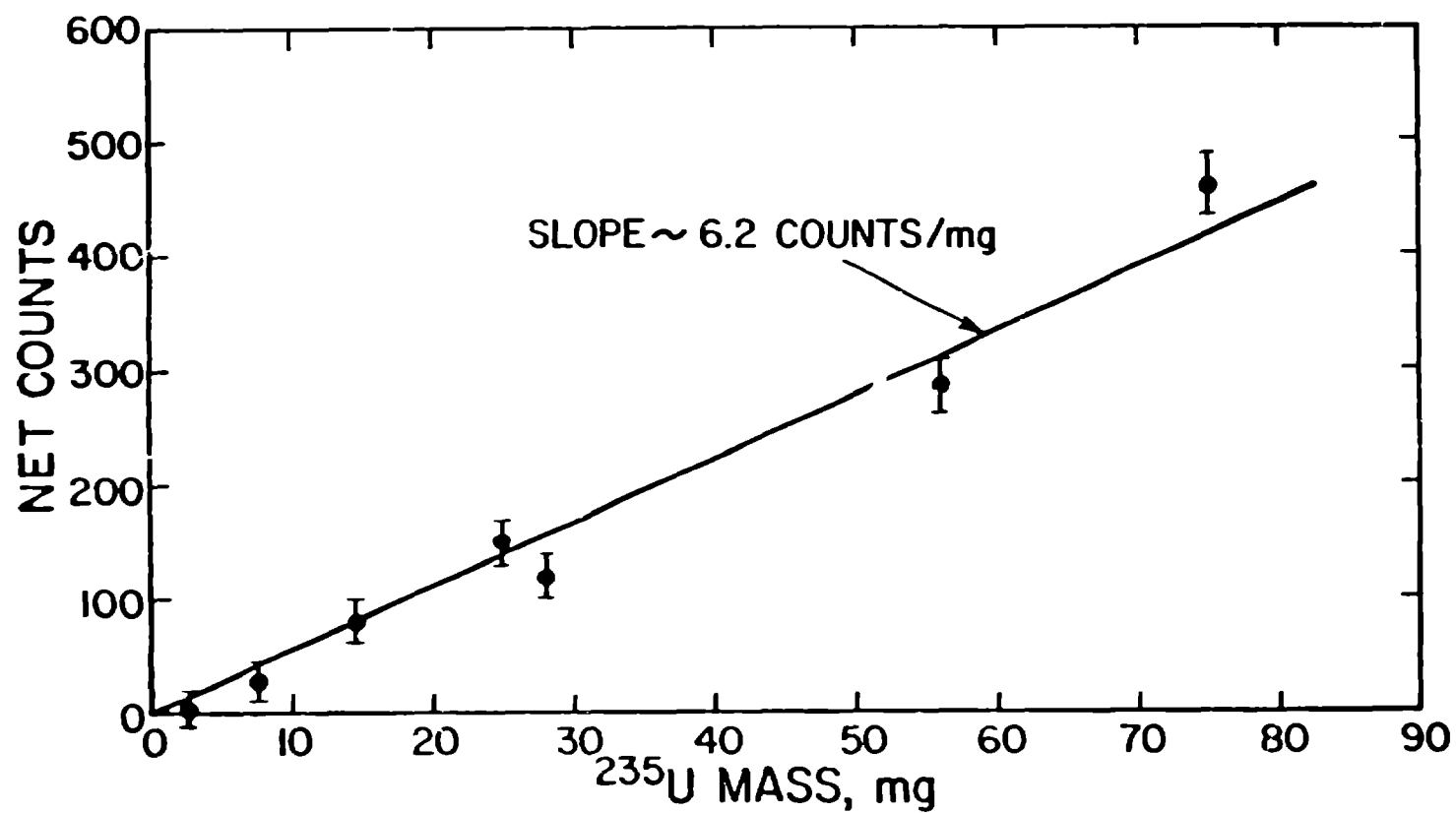


Fig. 3. Experimental response obtained with the prototype system shown in Fig 1. for 8000 interrogating pulses (80 s) from a 14 MeV neutron generator having a nominal 1×10^6 neutrons per pulse output. Low enrichment ^{235}U samples were placed at the center of a 200-liter barrel filled with sand for these measurements. The observed 3σ detection limit is 3 mg of ^{235}U .

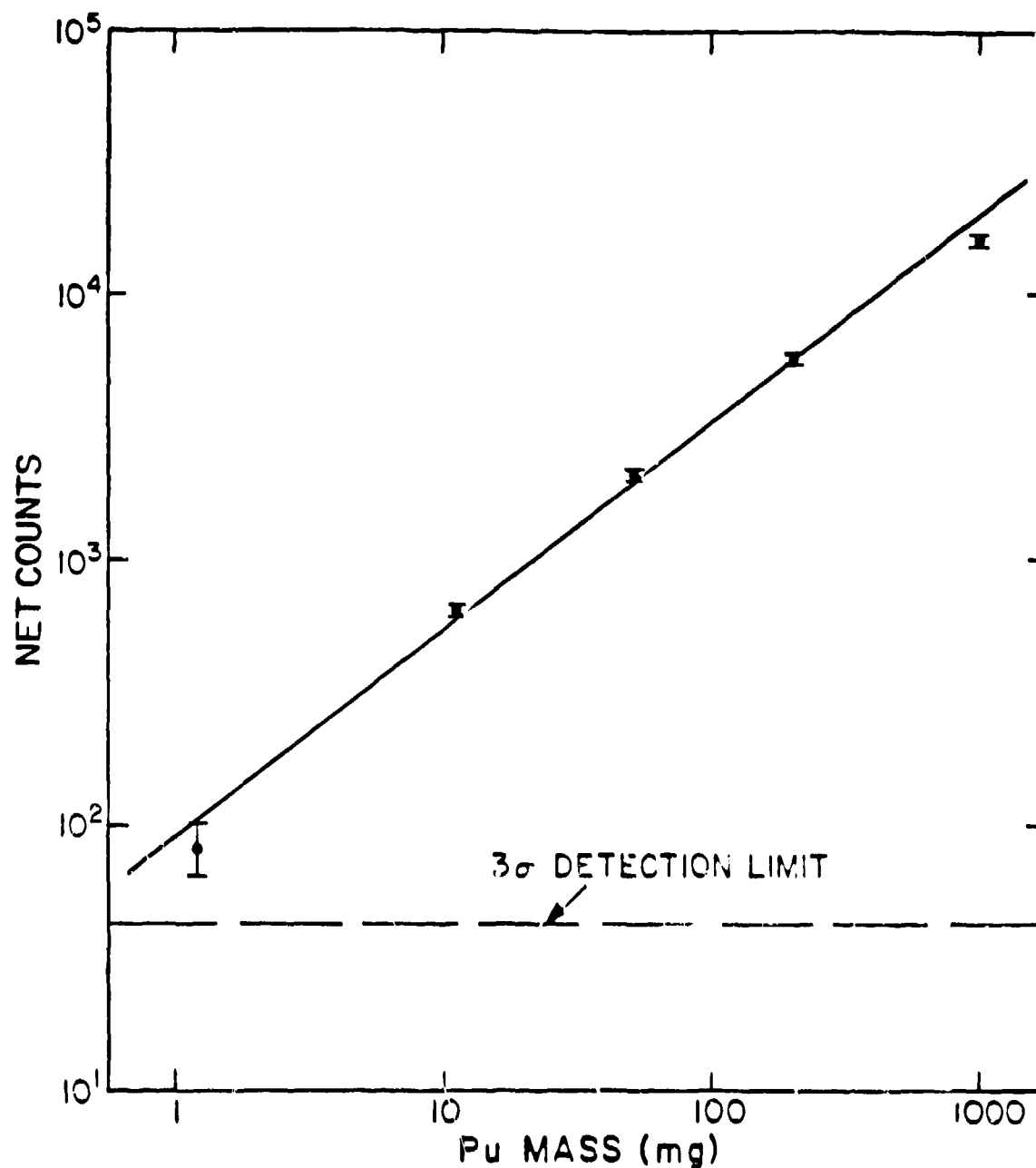


Fig. 4. Experimental response obtained in a series of 200-s runs with the Differential Dieaway Technique system (Fig. 1.) using pulsed $\text{Be}(\gamma, n)$ neutrons as the interrogating source. Data shown are for 20,000 electron linac pulses with about 4×10^6 $\text{Be}(\gamma, n)$ neutrons produced per pulse. Run total interrogating flux is about ten times that for the data shown in Fig. 3. Samples of PuO_2 were placed at the center of a 200-liter barrel of sand. The observed 3σ detection limit is 0.4 mg Pu (~ 0.2 nCi/g).